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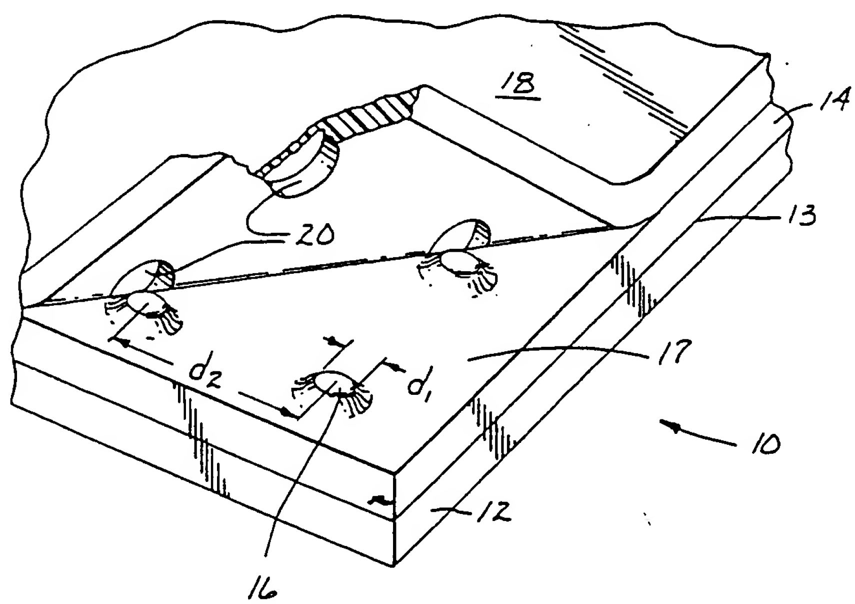

(57) Abstract

An adhesive sheet comprising (1) a backing, and (2) an adhesive layer having at least one topologically microstructured

Topologically structured adhesives have also been described. For example, relatively large scale embossing of an adhesive has been described to permanently reduce the PSA/substrate contact area and hence the bonding strength of the PSA (*See* EPO 0 279 579). Various adhesive layer topologies include concave and convex v-grooves or hemispheres, and other three dimensional shapes. In general, these topologies provide adhesive sheets, films and tapes with lower peel adhesion values in comparison with smooth surfaced adhesives. In many cases, the topologically structured adhesives also display a change in adhesion with increasing contact time.

The adhesive articles described herein above exhibit various types of positionable and repositionable behavior. Particles added to or applied over an adhesive to change its surface properties tend to alter the bulk adhesive properties. The articles generally suffer from problems of poor release liner attachment, poor aging, handling and bonding characteristics. In sign graphic applications, they tend to be difficult to print without destroying the positionable behavior of the construction. Furthermore, when adhesive articles are prepared using clear films and applied to clear substrates, particles (when pressed against the substrate) are large enough to be seen and result in visual imperfection, particularly, in backlit sign graphic applications.

Summary of the Invention

Briefly, in one aspect of the present invention an adhesive sheet is provided, having a backing and at least one topologically microstructured adhesive surface. The microstructured adhesive surface comprises a uniform distribution of adhesive pegs over the functional portion of an adhesive surface and protruding outward from the adhesive surface. The adhesive-only pegs typically comprise the same adhesive material as the underlying adhesive layer. The pegs have essentially flat tops that generally have a contact area of 1-25% of the total adhesive area. The pegs have an average height of at least 15 μm. A feature of the microstructured adhesive surface permits weak

Description of the Preferred Embodiment(s)

The present invention is an adhesive sheet having at least one topologically microstructured adhesive surface and a backing. The microstructured adhesive surface comprises a uniform distribution of adhesive pegs, protruding outward from the adhesive surface. The pegs generally comprise the same adhesive material as the underlying adhesive layer. The pegs have essentially flat tops that generally have a contact area of 1-25% of the total adhesive area. The pegs have an average height of at least 15 μm. The microstructured adhesive surface permits weak adherence of the sheet to a substrate, thus permitting easy repositioning as needed.

Advantageously, the microstructured adhesive surface also makes it possible to apply the sheet, such that a strong, permanent bond to the substrate is quickly established. The pegs provide repositionable adhesion with a light pressing on the adhesive sheet. Stronger adhesion can be made by compressing the pegs and contacting the underlying adhesive layer to the substrate. Advantageously, the microstructured adhesive surface of the present invention does not affect the aging characteristics of the adhesive or the backing, nor does it detract from the fully developed permanent bond.

Several embodiments of the present invention can be illustrated by reference to Figures 1-2 and 5-8. In contrast to the present invention, a prior art particle clump is illustrated in Figures 3 and 4.

Referring to Figures 1 and 2, an adhesive sheet 10 is illustrated comprising a backing 12, attached to an adhesive layer 14, wherein adhesive layer 14 comprises uniformly spaced topologically microstructured pegs 16 protruding above adhesive layer surface 17. Adhesive layer 14 is further overlaid with a liner 18. Liner 18 comprises uniformly spaced depressions 20. Typically, during fabrication of adhesive sheet 10, adhesive layer 14 is coated onto liner 18, wherein the adhesive mixture is allowed to flow into peg depressions 20. Backing 12 is then laminated to the planar surface 13 of adhesive layer 14. Although backing 12 is illustrated as a single ply, backing 12 can be comprised of multiple plies of thermoplastic materials, additional

coalesce. The dried or coalesced resin is then overcoated with an adhesive solution. Such a procedure forms the "beads" in situ.

Typically, the pegs can be formed by direct coating an adhesive onto a microstructured liner. In the case of composite pegs, the pegs can be formed by direct coating of adhesive onto a microstructured liner, in which small depressions have been previously filled with beads and optionally, a functionally sufficient amount of polymeric binder. Alternatively, the composite pegs can be formed with a first pass coating of an adhesive/bead slurry, followed by an adhesive only coating. The second, adhesive only coating could be a chemically different adhesive provided the second adhesive is sufficiently compatible with the first pass coating adhesive. This is generally necessary to keep the pegs on the adhesive (second coating) surface after the liner is removed during application.

A method for preparing the adhesive articles comprising composite pegs, comprises the steps: (a) preparing a slurry of beads; (b) coating the slurry onto an embossed liner to fill depressions in the embossed liner; (c) wiping the embossed liner to remove excess slurry; and (d) coating the filled embossed liner with an adhesive solution and; (e) allowing the adhesive solution to adsorb into and around the beads before drying. The adhesive solution can be a latex adhesive solution or a solution containing reactive monomers, that is polymerizable monomers as a diluent.

A preferred liner for an adhesive article of the invention is a low adhesion surfaced polymeric plastic film. The liners can be protective liners, release liners, and the like. When the adhesive article is made by the first of the above-outlined methods, the plastic film can be embossed to form the depressions. The embossable plastic film can be self-supporting, but a preferred liner is kraft paper, one or more surfaces of which has a thin, embossable polyethylene coating that is covered by a silicone release coating. Useful self-supporting plastic films include but are not limited to plasticized poly(vinyl chloride) and biaxially oriented poly(ethylene terephthalate) and polypropylene, all of which are economical and have good strength,

The adhesive may be substantially nontacky at room temperature if it becomes tacky at an elevated temperature at which it is to be used. Alternatively, the adhesive may be nontacky to the touch but aggressively tacky to other substrates. These adhesives are substrate specific and provides an appropriate adhesive bond between the substrate and the adhesive sheet.

Where thicker pressure-sensitive adhesive coatings are desired, it may be desirable either to apply multiple layers of the adhesive or to photopolymerize an adhesive in situ. For example, mixtures of monomeric alkyl acrylates, copolymerizable monomers, such as acrylic acid and optionally, polymers can be copolymerized by exposure to ultraviolet radiation to a pressure-sensitive adhesive state.

The adhesive article is useful for a variety of applications, such nonlimiting examples include decorative and informative markings on vehicles; signage; buildings; fascia; awnings; tapes, such as, transfer tapes, double-sided tapes, photographic and other layout tapes; masking for paint spraying; wallpaper; and the like.

Objects and advantages of this invention are further illustrated by the following examples, but the particular materials and amounts thereof recited in these examples, as well as other conditions and details, should not be construed to unduly limit this invention. All materials are commercially available except where stated or otherwise made apparent.

Examples

The adhesive resins used to prepare the adhesive solutions 1-3 and 5 contained acrylic copolymers that were prepared by free radical polymerization of isooctylacrylate and acrylic acid according to free radical polymerization techniques known to those skilled in the art, such as described in Odian, *Principles of Polymerization*, 3rd ed. John Wiley and Sons, New York, 1991. In the following examples, all parts are given by weight. Each of the adhesive solutions 1, 2 and 5 were coated within 24 hours of preparation.

seconds. The sheet was removed and immediately laid adhesive side down on the heated plate and allowed to rest for at least 10 seconds. The edge of the sheet was lifted and the sheet pulled laterally. If it slid freely it was rated a "1". If the sheet slid with resistance, but could be easily lifted off the surface it was rated "2". If the sheet did not slide but could be lifted and repositioned without damage, it was rated a "3". If the sheet stuck to the plate and could not be repositioned without damage, it was rated a "4".

Wet out area measurement

Samples were applied to cleaned glass plates as described above in the section on peel adhesion testing. The glass slides were illuminated from the edges and the adhesive viewed from above using a video camera equipped with a macro lens (4810 Series solid-state CCD camera, Cohu, Inc., San Diego, CA). Where the adhesive contacts the glass, a bright image was observed, non-contact areas were dark. Images of approximately 1 square inch were sampled using a computer-based image capture and analysis system (JAVA software from Jandel Scientific and PCVision digitizing computer/video interface card). The contact or % wet out areas were calculated as the ratio of bright image/total image. Values reported for % wet out area are the mean values of 4 sampling locations. The standard deviations for these values are approximately 2 % of total area.

Samples were also prepared as above and adhesive wet out areas estimated from optical microscopy photos using the measured images from above for visual comparison. Reflective lighting was used and the glass to adhesive contact areas were darker. These estimated contact values are shown as "~ %" wet out in the data tables.

Examples 1-12 and C1-C2

The examples 1 - 12 and C1 - C2 showed that adhesive films having the microstructured adhesive surface comprised of adhesive pegs could be temporarily applied and easily repositioned until a more permanent bond was

removed and the adhesive surface examined under a microscope. Each sample showed an adhesive surface that substantially replicated the surface of the release liner. Adhesion and wet out values obtained for these examples are shown in Table III.

Example 13

SCOTCHCAL brand, 3650 adhesive film was laminated to the film side of a sample from Example 3 and this composite was cut into 2.54 cm x 20 cm strips. The strips were aged ("baked") in the oven at 65°C under a 370 gram weight. Microscopic inspection of the aged adhesive surface showed a pattern of adhesive pegs protruding approximately 20 μm above a smooth adhesive surface. The adhesive pegs substantially replicated the liner surface. The adhesive surface and adhesion behavior were like Example 3.

Example C5

SCOTCHCAL brand, 3650 adhesive film was laminated to the film side of a sample from Example C1 and this composite was cut into 2.54 cm x 20 cm strips. The strips were aged ("baked") in the oven at 65°C under a 370 gram weight. Microscopic inspection of the aged adhesive surface showed a smooth surface. The adhesive surface and adhesion behavior were like Example C1.

Example 14

The release liner from a sample of Example C1 was removed and a sample of the embossed release liner from Example 3 was laminated in its place. SCOTCHCAL brand, 3650 adhesive film was then laminated to the film side of the above sample and this composite was cut into 2.54 cm x 20 cm strips. The strips were aged ("baked") in the oven at 65°C under a 370 gram weight to impart a microstructure to the adhesive surface. Microscopic inspection of the adhesive surface showed a pattern of partially formed adhesive pegs protruding above a smooth adhesive surface. The wet out

Examples 32-34 showed that the beads need not be glass and matching the refractive index of the adhesive and beads improved the optical properties of applied adhesive films. The clear film Examples 32-34 were indistinguishable for application and bonding characteristics from Example 27. When applied to a polycarbonate plate using a 2 kg roll down weight and illuminated either (a) from the back or (b) from the plate edges, Example 33 was optically clear under all illumination conditions, Example 32 was optically clear under back illumination and slightly hazy under edge illumination and Example 34 was hazy under all illumination conditions.

Example C13 showed filling an adhesive with glass beads significantly smaller than the adhesive coating thickness greatly reduced the adhesive tack and bond strength.

Examples 27-29 and 31-32

Examples 27-29, 31 and 32 were prepared according to the following general procedure. Adhesive solution 1 was coated onto a release liner to provide an adhesive layer that had a dry thickness of 30-40 μm. The liner had a square array pattern of depressions containing Potters 5000 SPHERIGLASS beads (from Potters Industries, Inc., Parisippany, NJ), the dimensions and frequency of the depressions for each sample are given in Table V. 60 μm thick vinyl film was laminated to the adhesive layer on the liner to give a pressure sensitive adhesive film for Examples 27 - 29 and 31. A 60 μm thick clear PET film was laminated to the adhesive layer on the liner to give a pressure sensitive adhesive film for Examples 32 - 34. The liner was removed and the adhesive surface examined under a microscope. Each sample showed an adhesive surface that substantially replicated the surface of the release liner. The pegs substantially replicated the liner surface depressions and were composed of glass beads in adhesive.

Electron micrographs of the adhesive surface showed composite pegs of glass beads in an adhesive matrix and the adhesive surface between the composite pegs was substantially free of beads.

Example 33

Plastic beads (3 μm average diameter crosslinked methyl methacrylate-vinyl acetate copolymer beads having a refractive index of 1.47, from 3M) were used to prepare a clear self adhesive film as in Example 32. Microscopic examination of the adhesive surface showed that adhesive pegs substantially replicated the liner surface. The beads were not visible using optical microscopy. Example 33 was optically clear under all illumination conditions when mounted on a polycarbonate plate.

Example 34

Potter's 5000 SPHERIGLASS beads (7 μm average diameter glass beds having a refractive index of 1.51) were used to prepare a clear self adhesive film as in Example 32. Example 34 was hazy under all illumination conditions when applied to a clear polycarbonate plate.

Example C13

Adhesive solution 1 (30 parts) was mixed with Potters 5000 SPHERIGLASS beads (70 parts) in ethyl acetate (140 parts) and then coated onto a smooth release liner to give an adhesive layer having a dry thickness of 30-40 μm. 60 μm thick vinyl film was laminated to the adhesive on the liner to give a pressure sensitive adhesive film. Electron micrographs of the adhesive surface showed the adhesive layer covered the glass beads like the composite pegs in Examples 27-30. When laid on a horizontal aluminum test plate, the sheet was easily slid over or lifted off the surface. When the panel was held in a vertical position, the adhesive sheet clung. Adhesion did not substantially increase with longer contact times, higher application pressure or heating.

Example 35-40 and C14

Examples 35-37 and C13 showed that other adhesives could be used to make the repositionable/permanent adhesive films. Example 37, like

Table II

Sample	Slide Rating	Peg Dimension density/height	Peel Adhesion 180 g Roll Down	Peel Adhesion 2 kg Roll Down	Wet Out on Glass 180 g Roll Down	Wet Out on Glass 2 kg Roll Down
		lpi/μm	kg/cm	kg/cm	%	%
1	3	85/30	0.5	1.3	7	94
2	3	85/24	0.6	1.3	10	98
3	3	85/20	0.6	1.3	11	100
4	4	85/15	0.9	1.3	4	96
C1	4	0/3	1.2	1.3	94	98
C2	4	0/1	1.4	1.4	95	99
5	3	65/30	0.6	1.3	2	98
6	3	65/20	0.5	1.3	3	100
7	4	65/15	1.0	1.4	4	95
8	3	40/36	0.5	1.3	7	93
9	3	40/25	0.6	1.2	4	96
10	3	40/20	0.5	1.2	6	89
11	3	40/17	0.5	0.9	3	86
12	4	40/13	0.8	0.9	6	88

Table VI

Sample (Adhesive)	Slide Rating	Peg Dimension density/height	Peel Adhesion 180 g Roll Down	Peel Adhesion 2 kg Roll Down	Wet Out on Glass 180 g Roll Down	Wet Out on Glass 2 kg Roll Down
		lpi/μm	kg/cm	kg/cm	%	%
35(4)	1	65/40	< 0.01	0.7	---	---
36(4)	1	65/27	< 0.01	0.7	~4	~95
37(4)	2	65/16	0.03	0.7	---	---
C14(4)	4	0/1	0.7	0.7	---	---
38(5)	2	85/20	0.2	1.0	~5	~97
39(5)	2	85/20	0.1	0.9	~3	~97
40(5)	2	85/20	0.3	0.8	~5	~98
26(5)	2	85/20	0.3	0.9	~8	~99
C10(5)	4	0/3	0.5	0.9	~90	~95

21. The double-sided adhesive sheet according to claim 18, further comprising a backing between the first and second adhesive layer and adjacent to the nonstructured surfaces of the first and second adhesive layers.

22. A double-sided adhesive article comprising an adhesive layer having a topologically microstructured surface on both surfaces of the adhesive layer, wherein the microstructured surface comprises a plurality of pegs, substantially uniformly distributed and protruding outwardly from the adhesive layer, wherein the pegs have essentially flat tops that comprise less than 25% of the total surface contact area of the adhesive layer, and the planar adhesive surface contiguous with the base and between the pegs is greater than 30% of the total adhesive layer.

23. A double-sided adhesive article comprising an adhesive layer having a topologically microstructured surface on both surfaces of the adhesive layer, wherein the microstructured surface comprises a plurality of composite pegs, substantially uniformly distributed and protruding outwardly from the adhesive layer, wherein the composite pegs comprise adhesive and one or more beads, wherein the pegs comprise an adhesive surface area of less than 25% of the total surface area of the adhesive layer.

24. A transfer tape comprising at least one topologically microstructured surface comprising a plurality of pegs, substantially uniformly distributed and protruding outwardly from the adhesive layer, wherein the pegs have essentially flat tops that comprise less than 25% of the total surface contact area of the adhesive layer, and the planar adhesive surface contiguous with the base and between the pegs is greater than 30% of the total adhesive layer.

25. A transfer tape comprising at least one topologically microstructured surface comprising a plurality of composite pegs, substantially uniformly distributed and protruding outwardly from the adhesive layer,

(d) coating the partially filled embossed liner with an adhesive solution;

(e) allowing the adhesive solution to adsorb into and around the beads before drying.

43. A method of preparing the adhesive article of claim 7, comprising the steps:

(a) coating a latex resin onto an embossed liner to partially fill the liner;

(b) wiping the embossed liner to remove excess latex resin;

(c) drying the resin to partially dry or coalesce the latex resin;

(d) coating the partially filled embossed liner with a latex adhesive solution;

(e) allowing the latex adhesive solution to adsorb into and around the beads before drying.

44. A reflective article according to claim 1, wherein the backing is reflective sheeting.

45. A reflective article according to claim 7, wherein the backing is reflective sheeting.

46. An adhesive article comprising an adhesive layer having a topologically microstructured surface comprising a plurality of adhesive-only and composite pegs, substantially uniformly distributed and protruding outwardly from the adhesive layer, wherein the adhesive-only pegs have essentially flat tops that comprise less than 25% of the total surface contact area of the adhesive layer, and the planar adhesive surface contiguous with the base and between the pegs is greater than 30% of the total adhesive layer, and the composite pegs comprise adhesive and one or more beads, wherein the

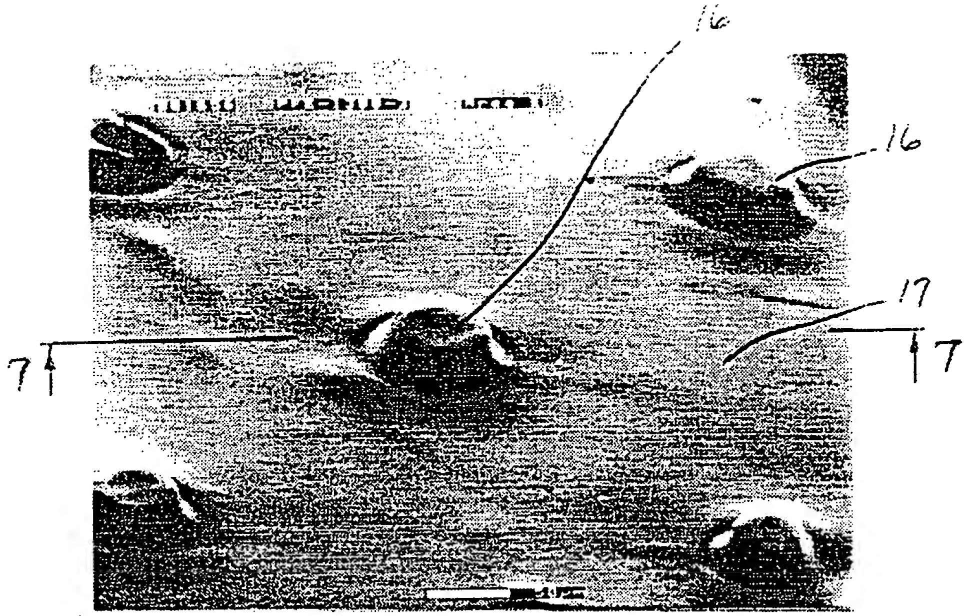

Fig. 2

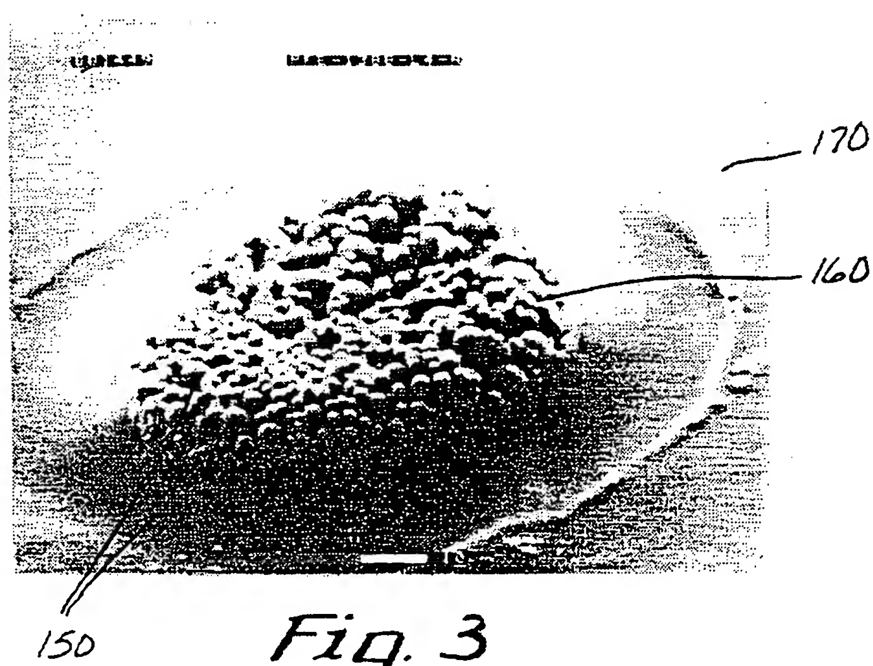

Fig. 3
PRIOR ART

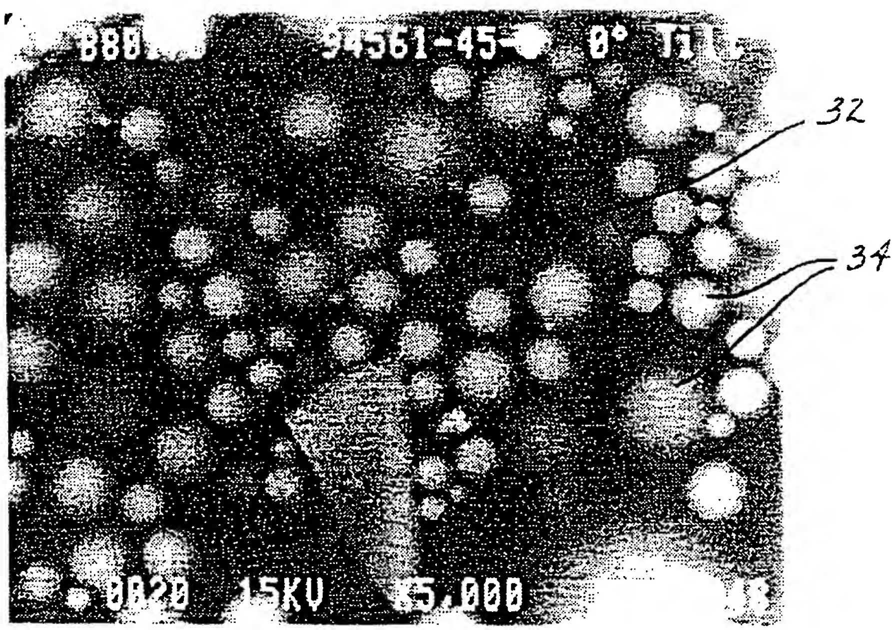

Fig. 8

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO.

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This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the European Patent Office EDP file on
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